

JUL 10 Rec'd PCT/PTO 15 JAN 2002

Form PTO-1390 U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE (REV 10-95) TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		ATTORNEY'S DOCKET NUMBER <b>4020-01239</b>
INTERNATIONAL APPLICATION NO. <b>PCT/EP00/06978</b>		U.S. APPLICATION NO. (37 CFR 1.55) <b>10/031054</b>
INTERNATIONAL FILING DATE <b>07.07.00 (07 July 2000)</b>	PRIORITY DATES CLAIMED <b>16.07.99 (16 July 1999)</b>	
TITLE OF INVENTION <b>RESIN FOR A MINERAL WOOL BINDER COMPRISING THE REACTION PRODUCT OF AN AMINE WITH A FIRST AND SECOND ANHYDRIDE</b>		
APPLICANT(S) FOR DO/EO/US <b>Erling HANSEN, Povel NISSEN, Thor HUSEMOEN, Dirk A. W. STANSSENS</b>		
<p>Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items And other information:</p> <p>1 <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371</p> <p>2 <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371</p> <p>3 <input checked="" type="checkbox"/> This expresses request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1)</p> <p>4 <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.</p> <p>5 <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))</p> <p>a <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau)</p> <p>b <input checked="" type="checkbox"/> has been transmitted by the International Bureau</p> <p>c <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US)</p> <p>6 <input type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2))</p> <p>7 <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))</p> <p>a <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau)</p> <p>b <input type="checkbox"/> have been transmitted by the International Bureau.</p> <p>c <input type="checkbox"/> have not been made, however, the time limit for making such amendments has NOT expired.</p> <p>d <input checked="" type="checkbox"/> have not been made and will not be made.</p> <p>8 <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</p> <p>9 <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4))</p> <p>10 <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5))</p> <p>Items 11. to 16. below concern document(s) or information included:</p> <p>11 <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98</p> <p>12 <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included</p> <p>13 <input checked="" type="checkbox"/> A <b>FIRST</b> preliminary amendment</p> <p><input type="checkbox"/> A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment</p> <p>14 <input type="checkbox"/> A substitute specification.</p> <p>15 <input type="checkbox"/> A change of power of attorney and/or address letter</p> <p>16 <input checked="" type="checkbox"/> Other items or information.</p> <p>a. WO 01/05725-Front Page, Specification And Claims (12 pp.)</p> <p>b. International Search Report (3 pp.)</p>		

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15 JAN 2002

U.S. APPLICATION NO. <b>107031054</b>		INTERNATIONAL APPLICATION NO. PCT/EP00/06978		ATTORNEY'S DOCKET NUMBER 4020-012139	
17. <input checked="" type="checkbox"/> The following fees are submitted <b>BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)):</b> Search Report has been prepared by the EPO or JPO <b>\$890.00</b> International preliminary examination fee paid to USPTO (37 CFR 1.482) <b>\$710.00</b> No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)) . . . <b>\$740.00</b> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO..... <b>\$1040.00</b> International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4)..... <b>\$100.00</b>				CALCULATIONS PTO USE ONLY	
ENTER APPROPRIATE BASIC FEE AMOUNT =				\$ 890.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input checked="" type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(c))				\$ 130.00	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	28 - 20	8	X \$18.00	\$ 144.00	
Independent claims	3 - 3 -	0	X \$84.00	\$ 0.00	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$280.00	\$ 0.00	
<b>TOTAL OF ABOVE CALCULATIONS =</b>				\$ 1164.00	
Reduction of 1/2 for filing by small entity, if applicable				\$ 0.00	
<b>SUBTOTAL =</b>				\$ 1164.00	
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$ 0.00	
<b>TOTAL NATIONAL FEE =</b>				\$ 1164.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) <b>\$40.00 per property</b> +				\$ 0.00	
<b>TOTAL FEES ENCLOSED =</b>				\$ 1164.00	
				Amount to be Refunded	\$
				Charged	\$
a. <input checked="" type="checkbox"/> A check in the amount of <b>\$ 1164</b> to cover the above fees is enclosed. b. <input type="checkbox"/> Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees A duplicate copy of this sheet is enclosed. c. <input checked="" type="checkbox"/> The Assistant Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>23-0650</u> A duplicate copy of this sheet is enclosed.					
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.					
SEND ALL CORRESPONDENCE TO Barbara E. Johnson 700 Koppers Building 436 Seventh Avenue Pittsburgh, Pennsylvania 15219-1818 Telephone: (412) 471-8815 Facsimile: (412) 471-4094					
SIGNATURE  _____ NAME Barbara E. Johnson _____ 31,198 REGISTRATION NUMBER					

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PATENT APPLICATION/PCT  
Attorney Docket No. 4020-012139

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re application of :  
  
Erling HANSEN : RESIN FOR A MINERAL WOOL BINDER  
Povl NISSEN : COMPRISING THE REACTION PRODUCT  
Thor HUSEMOEN : OF AN AMINE WITH A FIRST AND  
Dirk A. W. STANSSENS : SECOND ANHYDRIDE  
  
International Application :  
No. PCT/EP00/06978 :  
  
International Filing Date :  
07 July 2000 :  
  
Priority Date Claimed :  
16 July 1999 :  
  
Serial No. Not Yet Assigned :  
  
Filed Concurrently Herewith :

Pittsburgh, Pennsylvania  
January 15, 2002

**PRELIMINARY AMENDMENT**

Box PCT  
Commissioner for Patents  
Washington, D.C. 20231

Sir:

Prior to initial examination, please amend the above-identified patent  
application as follows:

**IN THE SPECIFICATION:**

**On page 1, after the title, please insert the following section heading:**

**BACKGROUND OF THE INVENTION**

1. Field of the Invention

**Before the paragraph beginning at page 1, line 10, please insert the following section heading:**

2. Description of the Related Art

**Before the paragraph beginning at page 1, line 24, please insert the following section heading:**

SUMMARY OF THE INVENTION

**Please replace the paragraph beginning at page 1, line 24 with the following rewritten paragraph:**

According to a first aspect of the present invention there is provided a resin for a binder suitable for mineral fibers such as glass or stone wool.

**Please replace the paragraph beginning at page 2, line 11 with the following rewritten paragraph:**

According to a second aspect of the present invention there is provided a binder.

**Please replace the paragraph beginning at page 2, line 14 with the following rewritten paragraph:**

According to a further aspect of the present invention there is provided a mineral fibre product.

**Please replace the paragraph beginning at page 2, line 17 with the following rewritten paragraph:**

According to yet a further aspect of the present invention there is provided a process for providing a polymer free resin for a binder suitable for binding mineral fibre products.

Before the paragraph beginning at page 2, line 24, please insert the following section heading:

DESCRIPTION OF THE PREFERRED EMBODIMENTS

IN THE CLAIMS:

Please cancel original claims 1-15 and rewrite them as new claims 16-43 as follows:

16. A resin for a binder suitable for mineral fibers such as glass or stone wool, said resin comprising the reaction product of a polymer free mixture of an amine with a first anhydride and a second anhydride, characterized in that the first anhydride and the second anhydride are different anhydrides.

17. The resin for a binder suitable for mineral fibers such as glass or stone wool according to claim 16, wherein the first anhydride is a cyclic anhydride and the second anhydride is a cyclic anhydride.

18. The resin for a binder suitable for mineral fibers such as glass or stone wool according to claim 17, wherein the first anhydride is an aliphatic anhydride and the second anhydride is an aromatic anhydride.

19. The resin according to claim 18, wherein the aliphatic anhydride comprises one or more anhydrides selected from the group consisting of tetrahydrophthalic anhydride, hexahydrophthalic anhydride, methyltetrahydrophthalic anhydride, succinic anhydride, nadic anhydride, maleic anhydride, and glutaric anhydride.

20. The resin according to claim 18, wherein the aromatic anhydride comprises one or more anhydrides selected from the group consisting of phthalic anhydride and trimellitic anhydride and/or pyromellitic dianhydride and methylphthalic anhydride.

21. The resin according to claim 18, wherein the concentration of aliphatic anhydride is greater than the concentration of aromatic anhydride.

22. The resin according to claim 16, wherein the amine is a N-substituted beta hydroxy alkylamine selected from the group consisting of ethanolamine, 1-ethylethanolamine, 1-methylethanolamine, n-butyl-ethanolamine, 1-ethylisopropanolamine, 1-methylisopropanolamine, 3-amino-1,2-propanediol, 2-amino-1,3-propanediol, tris(hydroxymethyl)aminomethane, and diethanolamine.

23. A resin comprising a polymer free mixture for a binder, said resin comprising the reaction product of a cyclic anhydride and an amine, at a pH of from about 2.5 to about 4.2, said pH being predetermined to positively influence the curing speed of the resin.

24. The resin according to claim 23, wherein the cyclic anhydride comprises a first anhydride which is an aliphatic anhydride and a second anhydride which is an aromatic anhydride.

25. The resin according to claim 24, wherein the aliphatic anhydride comprises one or more anhydrides selected from the group consisting of tetrahydrophthalic anhydride, hexahydrophthalic anhydride, methyltetrahydrophthalic anhydride, succinic anhydride, nadic anhydride, maleic anhydride, and glutaric anhydride.

26. The resin according to claim 24, wherein the aromatic anhydride comprises one or more anhydrides selected from the group consisting of phthalic anhydride and trimellitic anhydride and/or pyromellitic dianhydride and methylphthalic anhydride.

27. The resin according to claim 24, wherein the concentration of aliphatic anhydride is greater than the concentration of aromatic anhydride.

28. The resin according to claim 23, wherein the amine is a N-substituted beta hydroxy alkylamine selected from the group consisting of ethanolamine, 1-ethylethanolamine, 1-methylethanolamine, n-butyl-ethanolamine, 1-ethylisopropanolamine, 1-methylisopropanolamine, 3-amino-1,2-propanediol, 2-amino-1,3-propanediol, tris(hydroxymethyl)aminomethane, and diethanolamine.

29. The binder for mineral fibers such as glass or stone wool comprising the resin according to claim 16.

30. The binder according to claim 29, further comprising an accelerator and one or more resin additives selected from the group consisting of aminopropyl siloxane, thermal stabilizers, UV stabilizers, surface active compounds, fillers, silicates, magnesium sulfate, hydrophobising agents, oils, minerals, and silicone oils.

31. The binder according to claim 30, wherein the accelerator is selected from the group comprising sodium phosphinate, phosphinic acid, citric acid, adipic acid and g-hydroxyalkylamid.

32. The binder according to claim 29, further comprising one or more additives selected from the group comprising monosaccharides, disaccharides, and polysaccharides.

33. The binder according to claim 32, wherein the monosaccharides, disaccharides, and polysaccharides are one or more selected from the group consisting of sucrose, glucose syrup, modified starch, starch urea dicyandiamid, polyglycols, acrylics, furfural, carboxymethyl cellulose and cellulose, or polyvinyl alcohol.

34. The binder according to claim 29, wherein the binder has been cured.

35. The binder for mineral fibers such as glass or stone wool comprising the resin according to claim 23.

36. The binder according to claim 35, further comprising an accelerator and one or more resin additives selected from the group consisting of aminopropyl siloxane, thermal stabilizers, UV stabilizers, surface active compounds, fillers, silicates, magnesium sulfate, hydrophobising agents, oils, minerals, and silicone oils.

37. The binder according to claim 36, wherein the accelerator is selected from the group comprising sodium phosphinate, phosphinic acid, citric acid, adipic acid and g-hydroxyalkylamid.

38. The binder according to claim 35, further comprising one or more additives selected from the group comprising monosaccharides, disaccharides, and polysaccharides.

39. The binder according to claim 38, wherein the monosaccharides, disaccharides, and polysaccharides are one or more selected from the group consisting of sucrose, glucose syrup, modified starch, starch urea dicyandiamid, polyglycols, acrylics, furfural, carboxymethyl cellulose and cellulose, or polyvinyl alcohol.

40. The binder according to claim 35, wherein the binder has been cured.

41. The mineral fiber product bound by a cured binder according to claim 29.

42. The mineral fiber product bound by a cured binder according to claim 35.

43. A method for providing a polymer free resin for a binder suitable for binding mineral fiber products, said process comprising the steps of mixing together under reaction conditions an amine with a first aliphatic cyclic anhydride and a second aromatic cyclic anhydride.

#### **IN THE ABSTRACT:**

After the claims, please insert a page containing the Abstract Of The Disclosure, which is attached hereto as a separately typed page.



REMARKS

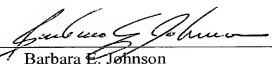
The specification and claim amendments have been made in order to conform this patent application to customary United States patent practice.

Attached hereto is a marked-up version of the changes made to the specification by the current amendment. The attachment is captioned "VERSION WITH MARKINGS TO SHOW CHANGES MADE".

Examination and allowance of pending claims 16-43 are respectfully requested.

Respectfully submitted,

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RESIN FOR A MINERAL WOOL BINDER COMPRISING THE  
REACTION PRODUCT OF AN AMINE WITH A FIRST AND  
SECOND ANHYDRIDE

ABSTRACT OF THE INVENTION

Resin for a binder suitable for mineral fibers such as glass or stone wool, said resin comprising the reaction product of a polymer free mixture of an amine with a first anhydride and a second anhydride, characterized in that the first and second anhydrides are different anhydrides.

**VERSION WITH MARKINGS TO SHOW CHANGES MADE****In the specification:**

**Paragraph beginning at page 1, line 24 has been amended as follows:**

According to a first aspect of the present invention there is provided a resin for a binder suitable for mineral fibers such as glass or stone wool [according to claims 1-6].

**Paragraph beginning at page 2, line 11 has been amended as follows:**

According to a second aspect of the present invention there is provided a binder [according to claims 7-12].

**Paragraph beginning at page 2, line 14 has been amended as follows:**

According to a further aspect of the present invention there is provided a mineral fibre product [according to claim 13].

**Paragraph beginning at page 2, line 17 has been amended as follows:**

According to yet a further aspect of the present invention there is provided a process for providing a polymer free resin for a binder suitable for binding mineral fibre products [according to claim 14].

RESIN FOR A MINERAL WOOL BINDER COMPRISING THE REACTION  
PRODUCT OF AN AMINE WITH A FIRST AND SECOND ANHYDRIDE

The invention relates to a compound or salts thereof suitable for use as a binder for mineral fibres, i.e. men made vitreous fibres (MMVF), for example glass slag or stone wool, i.e. mineral wool, in particular Rockwool, a binder composition comprising such a compound, a process for providing said compound and composition, a mineral fibre product provided with such a binder and the use of said compound and composition as a mineral fibre binder.

10 Phenol and formaldehyde resins which are mainly used as binders for glass or stonewool release toxic substances during curing, for example formaldehyde.

During application and curing of the binders, after provision thereof to the mineral fibres, phenol, 15 formaldehyde and ammonia are released. From an environmental point of view this is undesirable.

Furthermore during application, mostly by spraying, of the binder onto the spun glass or stone fibres a large amount of binder is lost, which is almost 20 impossible to recover for re-use.

A formaldehyde and phenol free resin suitable for a binder for mineral wool fibres is described in the patent application PCT/NL99/00029 from the applicant.

According to a first aspect of the present 25 invention there is provided a resin for a binder suitable for mineral fibres such as glass or stone wool according to claims 1-6.

The inventors have found that the resin obtained on mixing together different cyclic anhydrides, 30 preferably an aliphatic anhydride and an aromatic anhydride in a polymer free mixture with an amine,

provides a resin suitable for a mineral wool binder, which has desirable curing times.

The inventors theorize that the increased curing speed may not only be related to the combined use of cyclic anhydrides, but also to the adjustment of the pH, preferably to between 2.5 and 4.2, by employing aromatic carboxylic acids. These carboxylic acids are more acidic than aliphatic ones. Trimellitic anhydride is even more acidic due to the presence of an extra electron withdrawing group.

According to a second aspect of the present invention there is provided a binder according to claims 7-12.

According to a further aspect of the present invention there is provided a mineral fibre product according to claim 13.

According to yet a further aspect of the present invention there is provided a process for providing a polymer free resin for a binder suitable for binding mineral fibre products according to claim 14.

According to yet another use of the present invention there is provided the use of the above referred to resin in a mineral wool binder.

The invention will now be further clarified by way of the following description with reference to the examples.

Three binder compositions were made up according to the Mol.ratios as shown in table 1, whereby various accelerators were added to these compositions (see column 3 of table 1).

#### Example 1

##### Binder 1

A double jacketed glass reactor, heated with hot water, provided with a magnetic stirrer and a reflux condenser is used.

158 g diethanolamine (DEA) and 100 g water were brought into the reactor and heated to 70°C.

Then 228 g tetrahydrophthalic anhydride (THPA) was added allowing the temperature to raise to 90°C. This  
5 gives molratio DEA:THPA = 1:1

The reaction time was allowed to be 120 minutes before cooling of the resin.

Another resin with molratio DEA (158 g) : THPA (365 g) = 1:1,4 was made in the same way.

10 The binder solution was made by diluting the resin to about 40% solids with water. When accelerators were used they were added to the binder solution when it was ready for testing.

#### 15 Binder 2

The same equipment and procedure were used as for binder 1, except that phthalic anhydride (PTA) was added to the mixture when all THPA was dissolved.

3 resins were made with different molratios of  
20 the compounds.

Molratios used are:

DEA (158 g) : THPA (228 g) : PTA (89 g) = 1:1:0,4

DEA (158 g) : THPA (183 g) : PTA (133 g) = 1:0,8:0,6

25 DEA (158 g) : THPA (137 g) : PTA (178 g) = 1:0,6:0,8

The binder solution was made by diluting the resin to about 40% solids with water. When accelerators were used they were added to the binder solution when it  
30 was ready for testing.

#### Binder 3

The same equipment and procedure were used as for binder 1, except that trimellitic anhydride (TMA) was  
35 added to the mixture when all THPA was dissolved.

4 resins were made with different molratios of the compounds.

Molratios used are:

DEA (158 g) : THPA (297 g) : TMA (29 g) = 1:1,3:0,1

DEA (158 g) : THPA (274 g) : TMA (58 g) = 1:1,2:0,2

DEA (158 g) : THPA (228 g) : TMA (115 g) = 1:1:0,4

5 DEA (158 g) : THPA (183 g) : TMA (173 g) = 1:0,8:0,6

The binder solution was made by diluting the resin to about 40% solids with water. When accelerators were used they were added to the binder solution when it  
10 was ready for testing.

### Example 2

The curing characteristic of binders 1-4 were measured, the results of which are shown in table 1.

15 These show the reaction time at a certain temperature, measured as the time necessary to obtain a "not flowing" behaviour of the binder expressed as "flow time".

### Procedure:

20 3 droplets of binder solution (about 40% solids) are placed on a thin "Microscope glass cover slip" and dried in an incubator at 90°C for 45 minutes.

The "flow time" is measured by placing the pre-dried "microscope slip" on a metal plate with a pre-set  
25 temperature at 200°C. (Used is a Stork-Tronic Preziterm heating table.)

A very thin spatula or similar is used to stir the binder. When the binder becomes high viscous and sticks to the spatula, the time is measured as the "flow  
30 time".

5 samples are done, and by experience of the foregoing you wait as long as possible before start stirring to avoid cooling by the stirrer.

The average of the 2 last samples are used as  
35 the "flow time".

Table 1

	Composition of binder	Mol.ratios	Accelerator-% of solids	Flowtime minutes	Mechanical strength	
					Unaged	Aged
5						
	1) DEA:THPA	1:1	none	>10	4,80	2,55
			Citric acid-0,3-2%	5		
			Hypophos. acid-0,3-2%	5		
		1:1,4	none	>10		
10			Citric acid-3-4%	7		
			Hypophos.acid-1-2%	6		
			Na-phosphinat-4%	7		
	2) DEA:THPA:PTA	1:1:0,4	none	5	5,89	2,77
			Na-phosphinate-4%	7		
15			Hypophos.acid-1%	5		
		1:0,8:0,6	none	5	3,05	1,04
			Na-phosphinat-4%	7		
			Hypophos acid-1%	5		
		1:0,6:0,8	none	5		
20			Na-phosphinate-4%	7		
			Hypophos.acid-1%	5		



Table 1 [continued]

5	3) DEA:THPA:TMA	1:1,3:0,1	none	4	8,55	2,77
			Hypophos.acid-0,5-1%	4		
		1:1,2:0,2	none	4	6,88	2,07
			Hypophos.acid-0,5-1%	4		
		1:1:0,4	none	5	3,81	1,25
10			Hypophos.acid-0,2-1%	4		
			Na-phosphinate-4%	2		
		1:0,8:0,6	none	3	2,62	
			Hypophos.acid-0,5-1%	3		

## Conclusion:

15 Addition of aromatic anhydrides as PTA and TMA reduce the "flowtime" with a factor 2.

20 Accelerators as citric acid, phosphonic acid or sodiumphosphinate reduce the "flowtime" by a factor 2 when THPA is used as the only anhydride. Accelerators do not decrease the "flowtime" further when aromatic anhydrides are in the formulation. They themselves act as an accelerator.

Example 3 Storage stability of resin made at 90°C  
reaction temperature compared to 70°C

The preferred reaction temperature is 90 to 100°C using formulations containing up till 30% W/W water of the weight of anhydride. The water is mixed with the diethanolamine before the anhydride is added.

Using lower reaction temperature may cause precipitation if stored more than 2 weeks at solids higher than 75%.

10 If diluted at once with water to solids below 60% no precipitation is observed.

From a storage and transport point of view it is advantageous to have as high solids content as possible.

15

Example 4 Batch reactor

The reflux cooling is started.

20 40 kg melted diethanolamine (60°C) is pumped into the reactor.

The stirrer is started, 24 kg demineralized water is added and the temperature raised to 70°C.

25 80 kg tetrahydrophthalic anhydride is added portion wise over approximate 15 minutes, not allowing the temperature to exceed 95°C. The reactor is cooled if necessary.

The reaction temperature is held at 90-95°C for 15 minutes after all the anhydride is dissolved.

30 After 15 minutes the resin was cooled to room temperature.

The resin contained approximately 80% solids determined at 200°C.

35

Example 5 Continuous reactor

5 A double jacketed stainless steel tube reactor with a static mixer at the inlet is used. The temperature in the reactor is steered by cooling or heating with water.

Diethanolamin at 60°C and a water suspension of tetrahydrophthalic anhydride is pumped into the tube reactor and mixed when passing the static mixer. The  
10 temperature is raised and adjusted to 90-95°C. Diethanolamin and the water suspension of tetrahydrophthalic anhydride are pumped from separate tanks in pre-set ratio for the resin formulation.

15 The flow of the components are adjusted so that it takes approximate 20 minutes after the reaction mixture has passed the static mixer, and left the reactor.

Volume of the tube reactor is 33 litres.

20 Flow is approximately 100 kg per hour, or 20 kg diethanolamine and 80 kg suspension containing 40 kg tetrahydrophthalic anhydride in 40 kg water, all per hour.

When the reaction mixture leaves the tube  
25 reactor it may be diluted with water to 30% solids, cooled to room temperature and is ready for use.

Other additives as silane coupling agent, curing accelerators, hydrophilic or hydrophobic agent etc. may be added to the binder "on stream" after the  
30 reactor.

The invention is not limited to the above description; the requested rights are rather determined by the following claims.

## CLAIMS

1. Resin for a binder suitable for mineral fibers such as glass or stone wool, said resin comprising the reaction product of a polymer free mixture of an amine with a first anhydride and a second anhydride, 5 characterized in that the first and second anhydrides are different anhydrides.

2. Resin for a binder suitable for mineral fibers such as glass or stone wool according to claim 1, wherein the first anhydride is a cyclic, preferably an 10 aliphatic anhydride and wherein the second anhydride is a cyclic, preferably an aromatic anhydride.

3. Resin comprising a polymer free mixture for a binder, said resin comprising the reaction product of a cyclic anhydride and an amine, at a pH, preferably being 15 between 2.5 and 4.2, said pH being predetermined to positively influence the curing speed of the resin.

4. Resin according to any of the claims 1-3, wherein the aliphatic anhydride comprises tetrahydro phthalic anhydride, and/or hexahydrophthalic anhydride, 20 methyltetrahydrophthalic anhydride, succinic anhydride, nadic anhydride, maleic anhydride, glutaric anhydride.

5. Resin according to any of the claims 1 to 4 wherein the aromatic anhydride comprises phthalic anhydride and trimellitic anhydride and/or pyromellitic 25 dianhydride, methylphthalic anhydride.

6. Resin according to any of the preceding claims, wherein the amine, being a N-substituted beta hydroxy alkylamine, is selected from the group (di)ethanolamine, 1-(m)ethylethanolamine, n-butyl- 30 ethanolamine, 1-(m)ethylisopropanolamine, 3-amino-1,2-propanediol, 2-amino-1,3-propanediol, tris(hydroxymethyl)aminomethane, most preferably diethanolamine.

7. Binder for mineral fibres such as glass or stone wool, said binder comprising a resin according to any of the claims 1-6.

8. Binder comprising a resin according to claim 5 7 further comprising an accelerator and one or more resin additives such as aminopropyl siloxane to improve adhesion to glass, thermal and UV stabilizers, surface active compounds, fillers such as clay, silicates, magnesiumsulfate and pigments such as titanium oxide, 10 hydrophobising agents such as fluorine compounds, oils, minerals and silicone oils.

9. Binder according to claim 8 wherein the accelerator is selected from the group comprising sodium phosphinate, phosphinic acid, citric acid, adipic acid 15 and  $\beta$ -hydroxyalkylamid.

10. Binder according to claims 8 or 9 wherein the additives are selected from the group comprising mono-, di-, and polysaccharides, such as sucrose, glucose syrup, modified starch, starch urea dicyandiamid, 20 polyglycols, acrylics, furfural, carboxymethyl cellulose and cellulose, or polyvinyl alcohol.

11. Binder according to any of the claims 7-10 or resin according to any of the claims 1-6, wherein the concentration of aliphatic anhydrides is greater than the 25 concentration of aromatic anhydrides.

12. Binder for mineral fibers, particularly mineral wool comprising a resin according to any of the preceding claims 1-6, which has been cured.

13. Mineral fibre product, specifically a 30 mineral wool product, bound by a cured binder according to claim 12, or claims 7-11.

14. Process for providing a polymer free resin for a binder suitable for binding mineral fibre products, said process comprising the steps of mixing together 35 under reaction conditions an amine with a first anhydride and a second different anhydride, these preferably being an aliphatic, cyclic anhydride and a cyclic aromatic anhydride respectively.

15. Use of a resin according to any of the claims 1-6 in a binder according to claims 7-11.

## (12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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(54) Title: RESIN FOR A MINERAL WOOL BINDER COMPRISING THE REACTION PRODUCT OF AN AMINE WITH A FIRST AND SECOND ANHYDRIDE

(57) Abstract: Resin for a binder suitable for mineral fibers such as glass or stone wool, said resin comprising the reaction product of a polymer free mixture of an amine with a first anhydride and a second anhydride, characterized in that the first and second anhydrides are different anhydrides.

WO 01/05725 A1

# Declaration and Power of Attorney For Patent Application

## English Language Declaration

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

Resin For A Mineral Wool Binder Comprising The Reaction Product Of An Amine  
With A First And Second Anhydride

the specification of which

(check one)

☐ is attached hereto.

☐ was received on \_\_\_\_\_ as

Application Serial No. \_\_\_\_\_

and was amended on \_\_\_\_\_

(if applicable)

☒ was filed as PCT international application

No. PCT/EP00/06978

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and was amended under PCT Article 19 on \_\_\_\_\_

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I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the patentability of this application in accordance with Title 37, Code of Federal Regulations, §1.56(a).

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)

Priority Claimed

<u>99202343.2</u>	<u>European</u>	<u>16 July 1999</u>	<input checked="" type="checkbox"/>	<input type="checkbox"/>
(Number)	(Country)	(Day/Month/Year Filed)	Yes	No
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(Number)	(Country)	(Day/Month/Year Filed)	Yes	No
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I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:



(Application Serial No.)	(Filing Date)	(Status)
		(patented, pending, abandoned)
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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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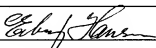
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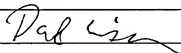
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